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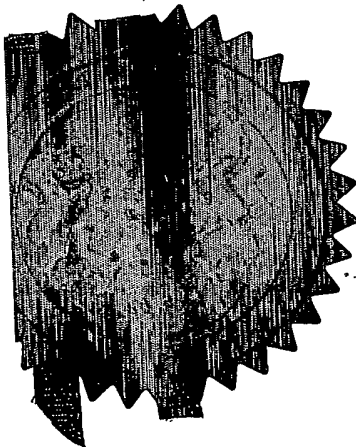
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- If the applicant is a corporate body, give the country/state of its incorporation GB
4. Title of the invention THERMAL INFRA-RED REFLECTIVE COATINGS

5. Name of your agent (if you have one) Bowdery Anthony Oliver
- "Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)
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Description 8

Claim(s) 2

Abstract 1

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DUPLICATE

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Thermal Infra-Red Reflective Coatings

5 This invention relates to the field of thermal energy control and more specifically to high thermal infrared (IR_T) reflective, paint formulations used in decorative coatings for inhabited buildings or other areas where control of IR_T energy and visual decoration are required.

Control of the spread of thermal energy in domestic buildings through passive techniques reduces energy consumption by reducing reliance on heating in cool environments and cooling air-conditioning in warm environments.

10 Convection as 'draughts' and conduction are widely recognised heat transport mechanisms and many materials and methods have been developed to reduce their effect on energy consumption in structures. Thermal energy may also be transported through radiative processes. Humans readily perceive conduction and convection processes in buildings but less so with IR_T energy. It is possible to control radiative energy transport processes to reduce or promote heat transfer into
15 a room using appropriate materials. For example metal reflectors, usually aluminium, are used behind radiators when mounted on the surfaces of outside walls and in cavity wall insulation materials where they control IR_T energy emitted from the surface. However the use of such reflective materials is currently restricted to areas where decorative appearance is not a primary concern, for example in loft spaces, or behind radiators.

20 All bodies above absolute zero emit radiation as a result of electronic processes in their atomic structure. At temperatures encountered in human habitations (0 to 30 °C) this electromagnetic radiation is predominately at frequencies in the range of 3×10^{12} to 3×10^{14} Hz, (1 to 100 microns wavelength equivalent) commonly referred to as the 'infrared region', and is caused by the vibration of electronic charge in the molecular structure of the material at these frequencies. At
25 temperatures in the range 0-30°C more than 90% of thermal energy is transported in the wavelength interval 3 to 50 microns, the Thermal Infrared (IR_T).

The energy emitted by a surface is given by the Stephan-Boltzmann equation:

$$W = \epsilon \sigma T^4 \quad Wm^{-2}$$

30 Where 'W' is the radiated power, 'ε' is the emissivity, 'σ' is the Stephan-Boltzmann constant and 'T' is the absolute temperature in Kelvin.

Emissivity (ε) is a property of the radiating surface material. Theoretically, emissivity can vary between 1 and 0. When ε=1 a surface will radiate with maximum efficiency and when emissivity=0 a surface does not radiate. Surfaces typically encountered in inhabited environments however, have emissivities over a smaller range. At general room temperatures (e.g. 0 to 30°C) and in the

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IR_T region organic materials tend to be highly absorbing and consequently have high emissivities, typically in the range 0.9 to 0.95. In practice the majority of decorative materials found in inhabited environments have organic surfaces, for example paints, timber, fabrics etc. and consequently have IR_T emissivities in the range 0.9 to 0.95.

- 5 Kirchhoffs law relates the emissivity of a material to its reflectivity and generally states that for opaque bodies:

$$\epsilon_{\lambda} = 1 - R_{\lambda}$$

Where ' ϵ_{λ} ' is the emissivity and ' R_{λ} ' the surface reflectivity at wavelength ' λ '. Thus, *reducing* the emissivity of an opaque surface *increases* its ability to *reflect* radiation. Consequently, an internal building wall with a low IR_T emissivity surface coating will reflect radiant heat from a heat source such as a radiator or an inhabitant, back into the room (which would be useful in cool climates). It also limits the radiation into a room from hot walls (which would be useful in hot climates).

The reflectivity of a conductive material is also related to its electrical resistivity. At IR_T frequencies, materials with metallic or near metallic conductivities have reflectivity given by the equation:

$$R = 1 - 2\sqrt{\nu\rho} + \dots$$

Where ' R ' is the reflectivity, ' ν ' is the frequency of radiation and ' ρ ' is resistivity. This predicts that metals with low electrical resistivity will have the highest reflectivity. Clean smooth conductive metals at temperatures between 0 and 30°C have high IR_T reflectivity, in the range 0.02 to 0.15.

- 20 Note: In the above equation the resistivity ρ is measured in units of ohm meter, Ωm . Resistivity measures the extent to which a substance offers resistance to passage of an electric current and the resistivity of a conductor in ohm meters is defined to be its resistance (in ohms) multiplied by its cross-sectional area (in square meters) divided by its length (in meters).

However, for surface films and other materials whose thicknesses are considered to be negligible resistivity is often quoted in "ohms per square", $\Omega\Box^{-1}$. The resistivity of a very thin conductor is defined to be its resistance (in ohms) multiplied by its width and divided by its length. If the conductor is square in shape, then its length and width are the same and its resistivity is numerically equal to the resistance of the square, which is actually the same no matter what the size of the square is. Therefore the resistivity could be stated in ohms, but it is conventional to state it in "ohms per square." One can consider the square to have sides equal to one unit, the size of the unit being immaterial.

So ohm-m is a measurement of bulk resistivity³ and "ohms per square" is for measuring sheet resistance. The relationship between the two measurements is: bulk resistivity = sheet resistance x thickness in m.

5 A conventionally decorated surface at 25°C with an emissivity of 0.95 in the IR_T will radiate up to 425Wm⁻² in the IR_T. This can be reduced to less than 90Wm⁻² using a coating with a low (0.2) IR_T emissive / high IR_T reflective / material surface.

10 A conventional, unmodified, paint typically comprises a mixture of colour pigments in a solvated, optically clear film-forming material known as the binder. The binder is conventionally an organic polymer in decorative coating for inhabited environments. Paints may also contain a wide range of additional materials such as flow improvers, wetting promoters etc., in small quantities. In addition to binding the colour pigments to the substrate, the binder also provides other desirable properties such as the gloss of the paint and corrosion or biological attack barrier.

A number of paint formulations having low IR_T emissivity (/high IR_T reflectivity) exist which are based on variants of conventional decorative paint fluids.

15 Type 1 IR_T reflective coatings are based on scattering pigments chosen and graded to try to maximise reflectivity in the associated frequency band. However, these formulations have relatively low reflectivity in the IR_T due to absorption in the pigment particles, multiple reflections and long path lengths through the organic polymer binder and poor scattering due to small differences in refractive index of the binders and conventional granular pigments at IR wavelengths. Reflectivities in the IR_T of greater than 0.3 are difficult to achieve in coloured coatings based on scattering granular pigments.

25 Type 2 high reflectivity paints have been developed that use metal flake pigment to provide the IR_T reflector component. Typically aluminium is used. Such metal-flake only paints can be formulated with usefully high reflectivities. A correctly formulated aluminium flake and binder-only paint system can be readily prepared with an IR_T reflectivity of between 0.85 and 0.9, using flakes that have a tendency to congregate and orientate at the surface (or leaf), and a binder chosen for high transparency in the IR_T waveband. The disadvantage of paints formed purely from metal flakes such as aluminium is aesthetic, since they appear silver-metallic in colour when in a low IR_T formulation (or grey-metallic in the case of metals such as tungsten). By using small metal flakes (30 (<5µm diameter) or rougherid flakes grey, non-metallic paints can be made, but there is a commensurate reduction in IR_T reflectivity due to scattering losses.

35 In type 3 paints, to achieve a coloured effect, the metal flakes are conventionally combined with visual pigments in the binder. When the paint has dried, a thin layer of the binder polymer loaded with the visual pigment is formed over the plate-like metal flakes to provide the visual colour, the 'colour layer'. When particulate visible pigments are added to flake-containing binders, the orientation of the flakes will be disturbed so that they no longer align with each other or with the

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surface of the paint. The mixture of (relatively large) high aspect ratio flakes and (relatively small and large agglomerates) low aspect ratio particles does not pack efficiently or regularly and the metal flakes become misorientated with the coating surface. The mis-alignment reduces the total IR_T reflectivity achievable with the paint system due to scattering related effects. A further
5 problem with type 3 paints is that the colouring pigments tend to increase the refractive index of the binder which increases reflection losses due to increased total internal reflection within the paint film. The thickness of the colour layer has to be controlled to retain IR_T transparency to allow the IR_T radiation to reach the reflector particles and be reflected back out. To achieve high IR_T reflectivity (>0.7), the colour layer thickness should be less than $10\mu m$. To achieve moderate
10 IR_T reflectivity (>0.5), the colour layer thickness should be less than $30\mu m$

The combined effects of the addition of low aspect ratio pigment particles to form coloured high IR_T reflective paints is to decrease the maximum reflectivity from around 0.85 for aluminum flake paints to 0.5 for light coloured paints, to 0.4 for medium shades and 0.3 for dark shades. .

15 The domestic/industrial durability of paints using a thin colour layer is limited since the removal of the thin colour layer through scuffing and abrasive cleaning etc. can reveal the presence of the aluminium layer causing the paint to appear 'silvery'.

It is therefore an object of the present invention to provide a high IR_T reflective additive for use in paint (and other coating) formulations that substantially mitigates or overcomes the problems associated with prior art formulations of types 1 to 3.

20 Accordingly this invention provides for an infrared reflective flake comprising an infrared reflective core flake and an infrared transparent material which is coated on some or all of the surface of the core flake (wherein the core has a thickness of less than 0.2 microns).

The flake according to the invention can be used as an additive in paint, gel, varnish and other coating formulations to provide a highly IR_T reflective coating.

25 The invention provides a composite material comprising a thin IR_T transparent layer which is adhered to a IR_T reflective core. Generally the IR_T transparent layer would be coated onto both sides of the flake core but for decorative effects only one side may be coated.

30 The IR_T transparent layer comprises a binder material with optionally a coloured material. This layer provides visual colour and mechanical strength together with chemical and environmental protection for the core material. The IR_T reflective core comprises either metallic or conductive oxide flakes.

35 By removing the dependence on granular high refractive index oxide pigments, as seen in the prior art systems, the misalignment of flakes and increased refractive index of the binder related reduction in IR_T of a coating using the new flakes is alleviated. Using low refractive index dyes for the colour option minimises any detrimental increase in binder refractive index.

5

The reflector layer needs to be sufficiently reflective to reflect the majority of incident IR_T . This equates to a DC electrical resistivity in the range 0.1 to $50\Omega\text{cm}^{-1}$ ideally in the range 1 to $10\Omega\text{cm}^{-1}$.

- 5 For a particular bulk resistivity, the reflector layer needs to be sufficiently thick to reflect the majority of incident IR_T radiation. However, the metal (metal oxide) core edge thickness should be minimised to reduce scatter and subsequent losses. Therefore, the core material should ideally be less than 0.2 microns thick.

- 10 For conductive metals such as an aluminium core material, the thickness is preferably in the range 0.016 to 0.05 microns thick and more preferably in the range 0.03 to 0.04 microns. For low conductivity metal such as a chromium based core, the thickness is preferably in the range 0.08 to 0.12 microns.

If the reflector core is rough it will scatter IR_T and reduce IR_T reflectivity. Therefore the core material should have a surface texture of less than 1 micron with a depth to pitch ratio of less than 0.5. Preferably the texture is less than 0.2 microns.

- 15 The reflective area of the flake core also has an effect on the IR_T reflectivity. If the reflector flakes are small compared to the wavelength of radiation then scattering losses become important. Preferably therefore the average diameter of the flake core is greater than 10 microns. Note: although the term "diameter" is used the skilled reader would understand that the flake does not need to be perfectly circular or disc-like and that irregular shaped flakes can be used. In the event
20 that the flake has an irregular shape the term diameter refers to the largest dimension of the flake.

Very large flakes become difficult to align during drying of the binder systems and so preferably the flake diameter is less than 100 microns. Flakes above around 50 microns become resolvable by the human eye and so more preferably for non-metallic effect decorative coatings, the flake diameter is in the range 10-50 microns and more preferably still in the range 30-40 microns.

- 25 The layer which covers the IR_T reflector core must be sufficiently transparent to transmit IR_T radiation through to the reflector core without significant loss. This covering layer can therefore conveniently comprise an organic polymer binder which is either non-polar or weakly polar. Examples include polymers of ethylene, butylene, propylene, acrylate, methacrylate and styrene or copolymers thereof. Colour can be introduced through the addition of visible band dyes chosen
30 for high specific absorption in the visible waveband, associated with electronic transitions, but weak specific absorption at IR_T wavelengths due to molecular vibration. Desirable dyes include azo, xanthene and anthraquinone dyes such as solvent yellow 94, orange 1 and red 24. In this way the visual, decorative colour requirements can be met without significant reduction of IR_T transparency. The radiation path length is minimised by using the minimum thickness of coloured
35 polymer layer commensurate with providing the required level of visual colour, mechanical strength and stability to processing. This requirement is met using a minimum thickness of 0.2

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micron. Reflector core coatings using organic polymer layer thickness in the range 0.2 to 2 microns is preferred.

Alternatively the coloured flake binder could be inorganic. Many inorganic materials will exhibit significant transparency in the IR_T wavelength range and some can be coloured using either dyestuffs, metal salts etc. or are naturally visible self-coloured. Examples of dyed or pigmented materials would include but are not limited to sol gel deposited materials such as oxides of silicon, titanium or aluminium. Examples of self-coloured, IR_T transparent materials include but are not limited to, semiconductor materials such as silicon and germanium.

In a variant of the present invention the IR_T reflective core can be made to be optically transparent. This is achieved by forming the core from a material that is transparent at visual wavelengths but remains reflective in the IR_T region. Examples of materials that become optically transparent in this way are very thin layers of silver, gold, copper or their alloys. Alternatively some optically transparent oxides have significant IR_T reflectivity, for example, but not limited to indium and fluorine doped tin oxides (ITO, FTO). In using these materials, an optically transparent coating can be formed that may be coloured if required using either the coloured organic binder or coloured inorganic coating described above. In this way a clear and, if required, colourless high IR_T reflective varnish can be made.

Optically transparent IR_T reflective metal cores will in general be less than 1nm thick. The actual thickness required is dependent on when it becomes sufficiently conductive. Cores become significantly IR_T reflective when their electrical resistivity is less than $10 \Omega \square^{-1}$.

Embodiments of the invention are described by way of example only with reference to the accompanying drawings in which

Figure 1 shows the form of a typical IR_T reflective flake according to this invention.

Figure 2 shows the difference in reflectivity in the wavelength range 2 to 20 microns between a TiO₂(Standard White) pigment and a typical visually coloured, high IR_T reflective pigment constructed according to the present invention. It can be seen that the high IR_T reflective pigment generally has a reflectance of approximately 90% compared to the standard pigment which varies greatly between 0 and 50% reflectance.

Figure 1 shows a typical flake formulation according to the invention. A flake 1 consists of a three layer structure (2, 4, 6). Layer 4 is the thin metallic core flake. Layer 4 is covered on either side by colour layers 2, 6 formed from a dyed or otherwise coloured organic binder. The colour layers 2, 6 provide mechanical strength and chemical and environmental protection to the core flake 4.

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The core flake 4 can be formed from a wide variety of metals or metallic oxides but for the case where it is formed from Aluminum it will typically have a thickness δ of between 30 and 40 nm. For a chromium core flake this thickness would be in the range 800 to 1200 nm.

5 The colour layers 2,6 typically will have a thickness of 200-2000nm and comprise an organic polymer binder with 1-20% wt/wt of a dye.

Examples of suitable polymer binders are acrylic resins such as poly methyl, ethyl or butyl acrylates. Suitable dyes include Azo metal complexes, phthalocyanines and anthroquinones.

10 A specific flake construction is a 800nm (± 100 nm) thick colour layer (layers 2&6 in Figure 1) formed from Elvacites 2041 polymer binder (produced by ICI Acrylics) with 10%wt/wt of Neozapon blue 807 (produced by BASF PLC) colour dye and a 35nm (± 5 nm) thick reflector layer (layer 4 in Figure 1) aluminium. For such a formulation the flake has an emissivity in the range 0.15 – 0.2 and a dark blue colour.

A flake according to this invention can be formed in a variety of ways but a preferred method of manufacture is described as follows.

15 A 25 μ m thick sacrificial web of polyethylene terephthalate polymer film is coated with a release layer of caruba wax on one or both sides. A layer of dye-containing polymer is then deposited onto treated surfaces of the sacrificial web by either free-flow coating or gravure printing processes and cured.

20 A layer of aluminium is then deposited onto the surface of the dye-containing polymer by evaporation. Finally a second layer of coloured polymer is deposited onto the evaporated aluminium again by a free flow coating or gravure printing process. This second polymer layer is again cured.

25 The coated three-layer material is then removed from the web, by for example, immersion in hot water. The caruba wax release layer melts and the IR_T reflective material can be recovered by filtration or floatation methods.

Finally the three-layer material can be reduced to appropriate sized flakes through conventional processing, for example, wet grinding. Other processes may include, for example, solution deposition onto suitable size flakes or defining flakes on the web before removal.

30 Related permutations of the basic structure that will create different decorative effects will be apparent to the skilled reader. For example, combinations of colourants can be used on each side to extend the number of colours available, different colourants or combinations of colourants can be used on each side, the intensity of shade on each side can be varied independently. The reflective metal can be exchanged for a visually coloured metal, for example copper or tungsten. The metal reflector can be textured in either periodic or a-periodic structures to add visual

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diffraction effects or controlled diffuse reflections respectively. This could be achieved by embossing the first coloured polymer film before deposition of the metal reflector layer. Clearly these permutations can be used in various combinations to create further decorative effects.

CLAIMS

1. An Infrared (IR_T) reflective flake comprising an infrared reflective core flake and an infrared transparent material which is coated on some or all of the surface of the core flake wherein the reflector core has a thickness of less than 0.2 microns.
- 5 2. A flake as claimed in claim 1 wherein the core has a DC electrical resistivity in the range 0.1 to 50 Ωcm^{-1} .
3. A flake as claimed in claim 1 wherein the core is aluminium and has thickness in the range 0.03 to 0.04 microns.
4. A flake as claimed in claim 1 wherein the core is chromium and has a thickness in the range
10 0.08 to 0.12 microns.
5. A flake as claimed in any preceding claim wherein the core flake has a surface texture of less than 1 micron and a depth to pitch ratio of less than 0.5.
6. A flake as claimed in claim 5 wherein the texture is in the range 0.2 to 0.4 microns.
7. A flake as claimed in any preceding claim wherein the flake has a diameter of 10 to 100
15 microns.
8. A flake as claimed in claim 7 wherein the diameter is in the range 10 – 50 microns.
9. A flake as claimed in claim 8 wherein the diameter is in the range 30 – 40 microns.
10. A flake as claimed in any preceding claim wherein the infrared transparent material comprises a non-polar or weakly polar organic polymer binder containing a coloured dye.
- 20 11. A flake as claimed in claim 10 wherein the infrared transparent material thickness is in the range 0.2 to 2 microns.
12. A flake as claimed in claims 1-11 wherein the infrared transparent material comprises an inorganic binder.
13. A flake as claimed in any preceding claim wherein the core has a thickness of less than 1 nm.
- 25 14. A flake as claimed in claim 13 wherein the DC resistivity is less than 10 Ωcm^{-1} .
15. A flake as claimed in claims 10, 11 or 12 wherein the core comprises indium or fluorine doped tin oxide.
16. A flake as claimed in any of claims 13 to 15 wherein the transparent covering material completely encapsulates the core material.

17. A paint formulation comprising infrared reflective flakes according to claims 1-15.

18. A varnish formulation comprising infrared reflective flakes according to claims 1-16.

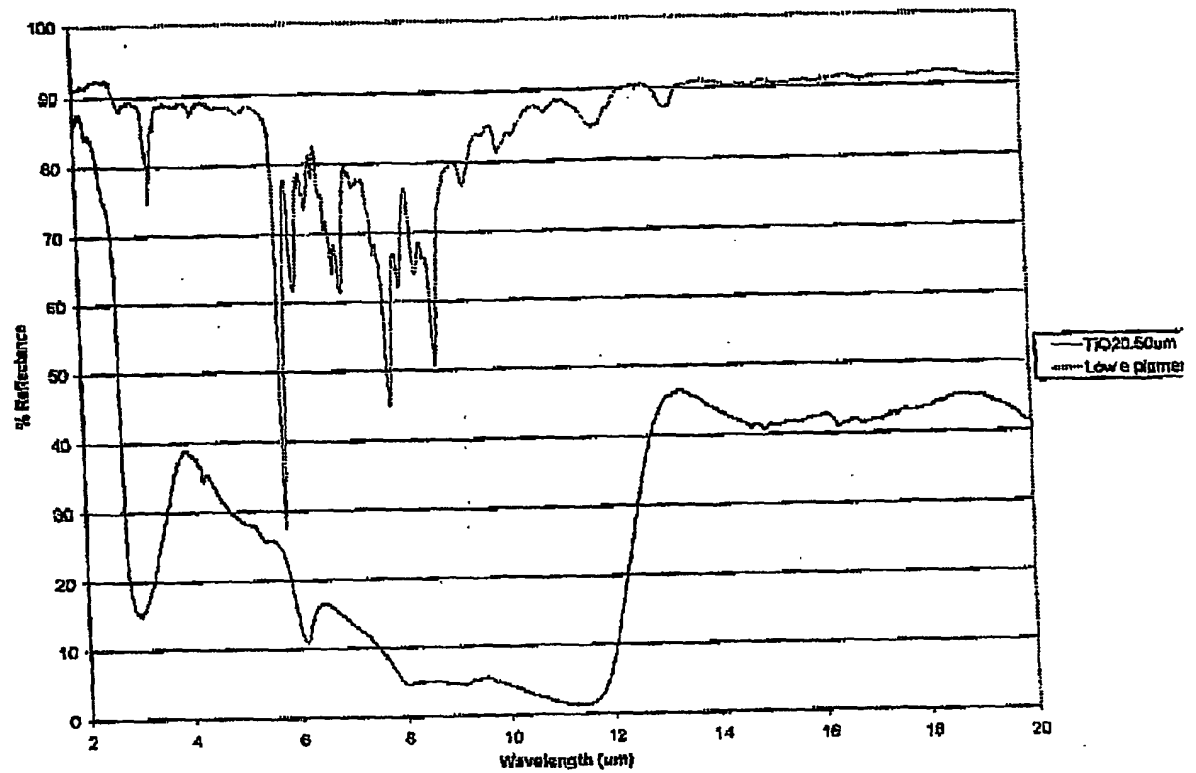
ABSTRACT

This invention relates to the field of thermal energy control and more specifically to low emissivity, infra-red reflective paint formulations. Control of the spread of thermal energy in domestic buildings through passive techniques reduces energy consumption by reducing reliance on heating in cool environments and cooling air-conditioning in warm environments. A number of paint formulations having low emissivity in the thermal infrared exist which are based on variants of conventional decorative paint fluids. There are a number of problems associated with such formulations such as susceptibility to damage and high emissivity for certain colour pigmentations. The present invention proposes a low emissivity flake for use in a paint formulation which substantially overcomes the problems associated with the prior art.

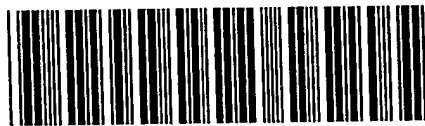
[Figure 1]

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Figure 2



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